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ORIGINAL ARTICLE

Organic matter enrichment in the first member of the Xiagou formation of the lower Cretaceous in the Jiuquan Basin, China

Lixin Pei[1] · Gang Gao[1] · Wenzhe Gang[1] · Zhilong Huang[1] · Guofu Ma[2] · Zhiming Yang[2] · Jianjun Chen[2]

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Abstract The sources and enrichment of organic matter in a sediment core in the first member of the Xiagou Formation (K_1g^1) from the Chang 2-2 borehole of the Jiuquan Basin, NW China, have been examined using Rock–Eval, maceral, carbon isotopes and biomarker data. This data indicates that highly variable organic matter sources and preservation conditions in response to climate change. TOC content, HI, and $\delta^{13}C$ value were strongly correlated with the abundance of gammacerane, woody organic matter content, steranes/hopanes ratio, and C_{29} sterane content. This correlation demonstrates the importance that the control of the salinity of the depositional environment and organic matter sources can have upon the enrichment, type, and carbon isotopic composition of organic matter. In the Jiuquan Basin's relatively high temperature and arid climate, high salinity lakes with high primary productivity of algae, planktons, and bacteria, and good organic matter preservation conditions (anoxic bottom water) resulted in the enrichment of isotopically-light algae-bacterial organic matter. In the Jiuquan Basin's regions with a relatively low temperature and wet climate, fresh lakes with low primary productivity of algae, planktons, and bacteria received significant terrigenous high plants input, resulting in the deposition of a low abundance of isotopically heavier terrestrial organic matter.

✉ Gang Gao
gaogang2819@sina.com; greathoper@163.com

[1] College of Geosciences, China University of Petroleum, Beijing, China

[2] Research Institute of Exploration and Development, Yumen Oilfield Company, CNPC, Jiuquan, Gansu, China

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1 Introduction

The Jiuquan Basin is a petroliferous basin in China. It contains a series of lacustrine sediments deposited during the early Cretaceous. This succession of lacustrine sediments preserves abundant paleoenvironmental information, which is important for understanding the mechanism of organic matter deposition in response to regional continental climate evolution. A considerable amount of studies on lake systems have resulted in variable lacustrine source rock models, such as the hypersaline lake model (Kirkland and Evans 1981), the large deep anoxic lake model (Demaison and Moore 1980), the oligotrophic mesosaline alkaine closed lake model (Kelts 1988), the meromictic/oligotrophic tropical/humid lake model (Talbot 1988), and the moderately deep tropical lake model (Katz 1990). Numerous studies of organic matter deposition in relation to Cretaceous climate and environments are based on marine sediments (Jenkyns 1980; Barron 1983; Arthur et al. 1988; Huber et al. 2002; Wagner et al. 2008; Wagreich et al. 2011), but there are few published studies regarding the Cretaceous continental environments and the related organic matter enrichment mechanisms (Hasegawa 1997; Hasegawa et al. 2003; Song et al. 2013). In the Jiuquan Basin, two major sets of organic rich source rocks developed during the early Cretaceous: the Chijinpu (K_1c) and the Xiagou (K_1g^{1+2+3}) formations. The lacustrine source rocks associated with the Chijinpu and the Xiagou Formation, respectively, were deposited in an arid to semi-arid climate based on the fossil plants and sediments (Pan et al. 2006; Deng and Lu 2008). Previous organic geochemical studies on the source rocks of the K_1c and the third member of the K_1g formation (K_1g^3) were used for oil-source correlation based on discontinuous core and outcrop mudstone samples (Xiong et al. 2006; Han et al. 2007). To date, there is no systematic geochemical study on the mechanism of organic matter enrichment in response to the climate evolution in the K_1g^1 (member 1).

The drilling of a continuous core profile from the K_1g^1 member in the Chang 2-2 borehole provides an opportunity for carrying out a systematic study of the source variations of organic matter and the depositional and environmental conditions. It also enables us to analyze the mechanism of organic matter deposition in the Jiuquan Basin. A comprehensive study of maceral, carbon isotopic composition of organic matter, and biomarkers and their variation along the profile has been undertaken. The purpose of this study is to analyze the mechanism of lacustrine organic matter enrichment in response to the climate evolution during Early Cretaceous in the Jiuquan Basin.

2 Sample collection and analytical methods

The Jiuquan Basin is located in northwestern China, and it covers an area of 22,000 km^2 (Pan et al. 2006) (Fig. 1). The Chang 2-2 borehole is situated on the Yinger Sag in the east of the Jiuquan Basin. The core samples used in this study were collected from a depth 3900 to 3990 m in the Chang 2-2 borehole, for the most part covering the K_1g^1 member. The lithology of sediments in this section is mainly grey to black mudstones, laminated mudstones, and silty mudstone.

All rock samples were cleaned with water and then dried prior to crushing and powdering. Rock–Eval analysis was performed on OGE-II according to China National Standard GB/T18602-2001.

Rocks were Soxhlet extracted using chloroform:methanol (87:13) for 72 h, and the isolated extractible organic matter was separated into saturated hydrocarbons, aromatic hydrocarbons, and polars on a silica:alumina column using hexane, hexcane: dichloromethane (2:1 v:v) and dichloromethane:methanol (1:1 v:v), respectively. Saturated hydrocarbons were analyzed for biomarkers. The extracted samples were treated with HCl and HF/HCl to separate kerogen from rock minerals. The kerogen concentrates were subjected to carbon isotope and maceral analysis.

Kerogen maceral composition analysis was carried out on a MPV-3 microscope-photometer. Kerogen is made into massive/powder brick polished sections and tested under oil-immersed reflection light. The organic maceral ratio determines fluorescent substances (mineral bituminite and exinite) and non-fluorescent substances under the induced blue light, and non-fluorescent substances are further determined to be either mineral or other organic matter components (vitrinite and inertinite) under common oil-immersed reflection light. The common oil-immersed reflection light conditions are objective lens $\times 50$ and eye lens $\times 10$, while the reflection fluorescence conditions are exciting filter disc K510, objective lens $\times 50$, and eye lens $\times 10$. Using induced blue light with oil-immerse reflection light, 500 points are counted to determine the composition of organic components.

Carbon isotope analysis of Kerogens was performed on a FLASH HT EA-MAT 253 IRMS according to China National Standard GB/T 18340.2-2001. Testing conditions were as follows: Carrier He (99.999 %), Flow 100 ml/min, Reference 250 ml/min, Combustion Gas Oxygen (99.995 %), Flow 250 ml/mim, Reactor Temp 980 °C, Reactor filling Cr_2O_3, reduced copper, and Ag/Cobalt Oxide.

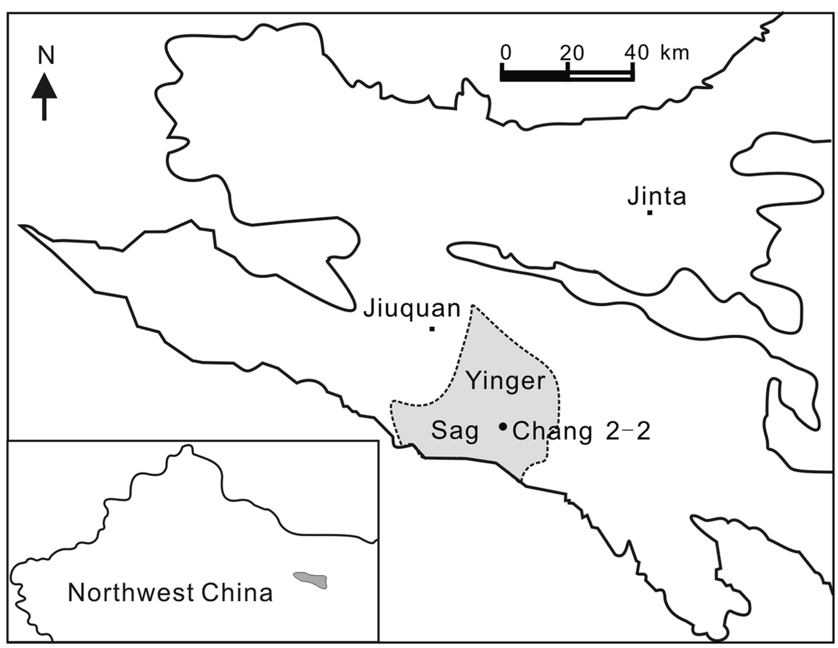

Fig. 1 Location of the Jiuquan Basin and the Chang 2-2 borehole

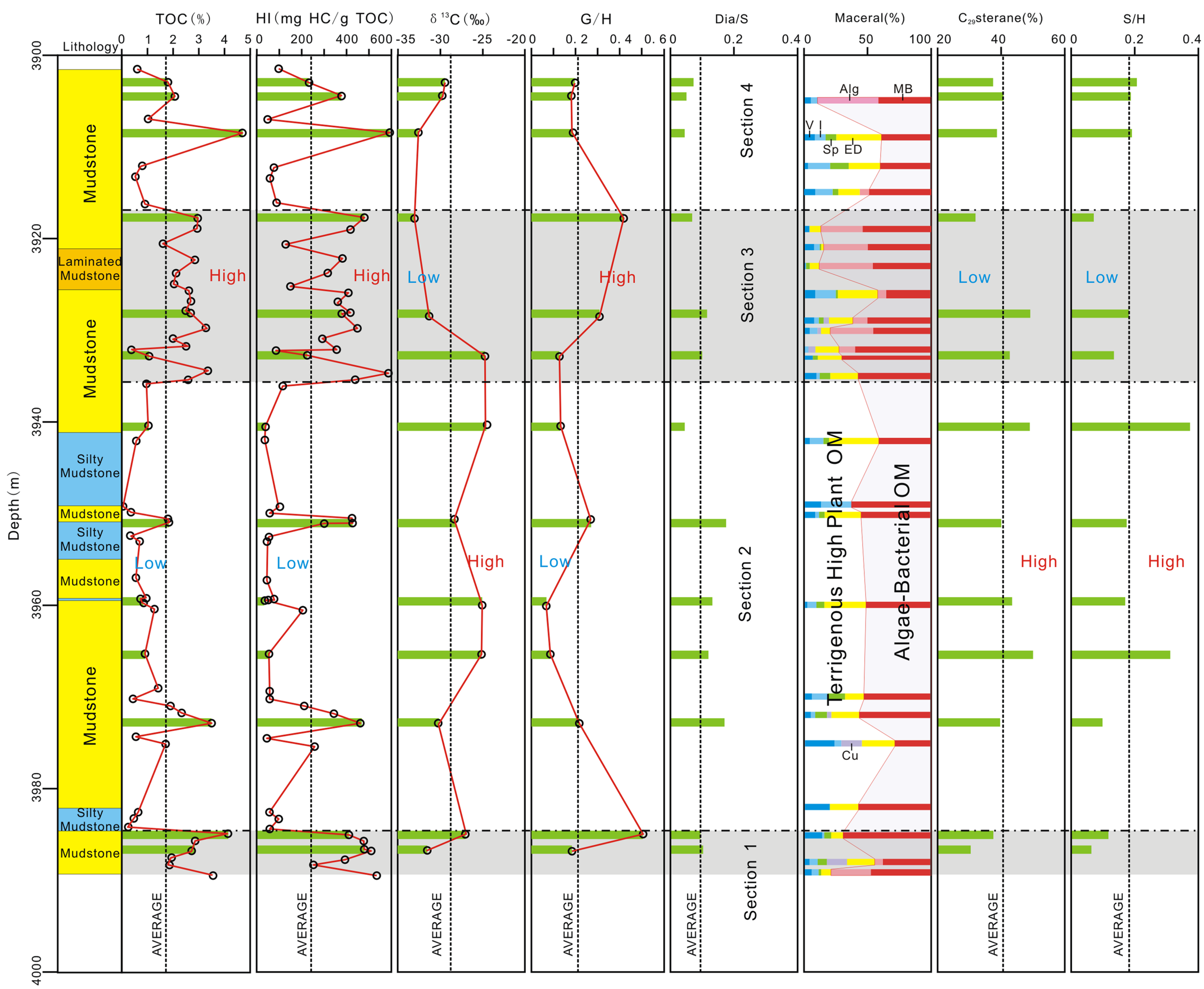

Fig. 2 Variation of the K_1g^1 member bulk, organic carbon isotopes and biomarker parameters with depth in the Chang 2-2 borehole. *G/H* gammacerane/C_{30} hopane, *Dia/S* C_{27-29} diasteranes/C_{27-29} steranes, *S/H* C_{27-29} steranes/C_{27-35} hopanes, *C_{29} sterane* (%) C_{29} sterane/C_{27-29} steranes, *OM* organic matter, *V* vitrinite, *I* inertinite, *Sp* sporinite, *Cu* cutinite, *ED* exinite debris, *Alg* alginite, *MB* mineral bituminite

GC–MS analysis of the saturate fractions was performed with a Thermo-Finnigan Trace-DSQ instrument equipped with a HP-5MS fused silica column (60 m × 0.25 mm × 0.25 m). The GC oven temperature for analysis of the saturate fractions was initially held at 50 °C for 1 min and then programmed to 120 °C at 20 °C/min, 250 °C at 4 °C/min, and 310 °C at 3 °C/min, and was finally held at 310 °C for 30 min.

3 Results and discussions

3.1 Abundance and types of organic matter

The total organic carbon content (TOC) for the K_1g^1 member samples from the Chang 2-2 borehole are depicted in Fig. 2. According to the relative content of TOC with respect to the average TOC, the K_1g^1 profile can be divided into four sections. Section 1 (mudstones) from 3989.30 to 3984.50 m has a high TOC content (1.80–4.12 %). Section 2 (mudstones and silty mudstones) from 3984.50 to 3935.50 m has the lowest TOC content (0.06–3.51 %, average 1.04 %) but includes a few high TOC intervals. Section 3 (mudstones and laminated mudstones) from 3935.50 to 3917.00 m is an organic-rich section with TOC content varying from 0.37% to 3.37 % (average 2.36 %). Section 4 (mudstones) from 3917.00 to 3901.50 m has a lower TOC content (0.51%–4.70 %, average 1.54 %).

The hydrogen index (HI) varies from 38 to 597 mg HC/g TOC and shows similar variation as that of the TOC contents (Fig. 3). As depicted in Fig. 3, the types of

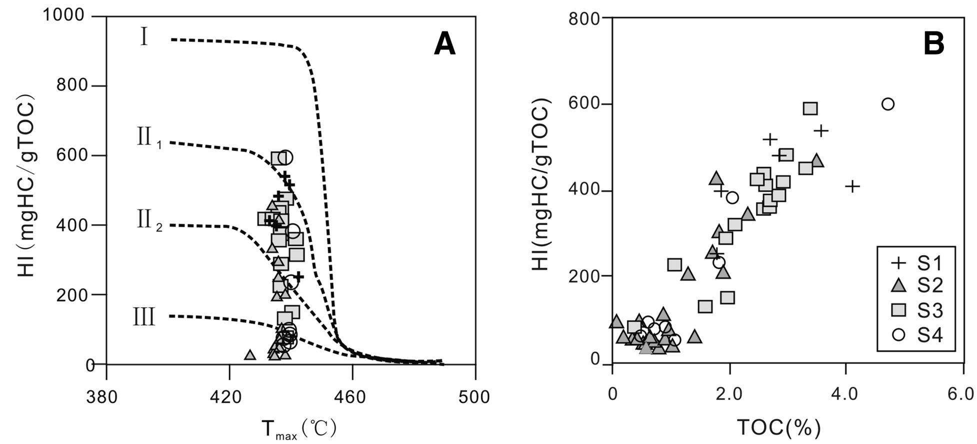

Fig. 3 Variation of Rock–Eval hydrogen index with Tmax (**a**) and TOC (**b**) for the K1g1 member samples

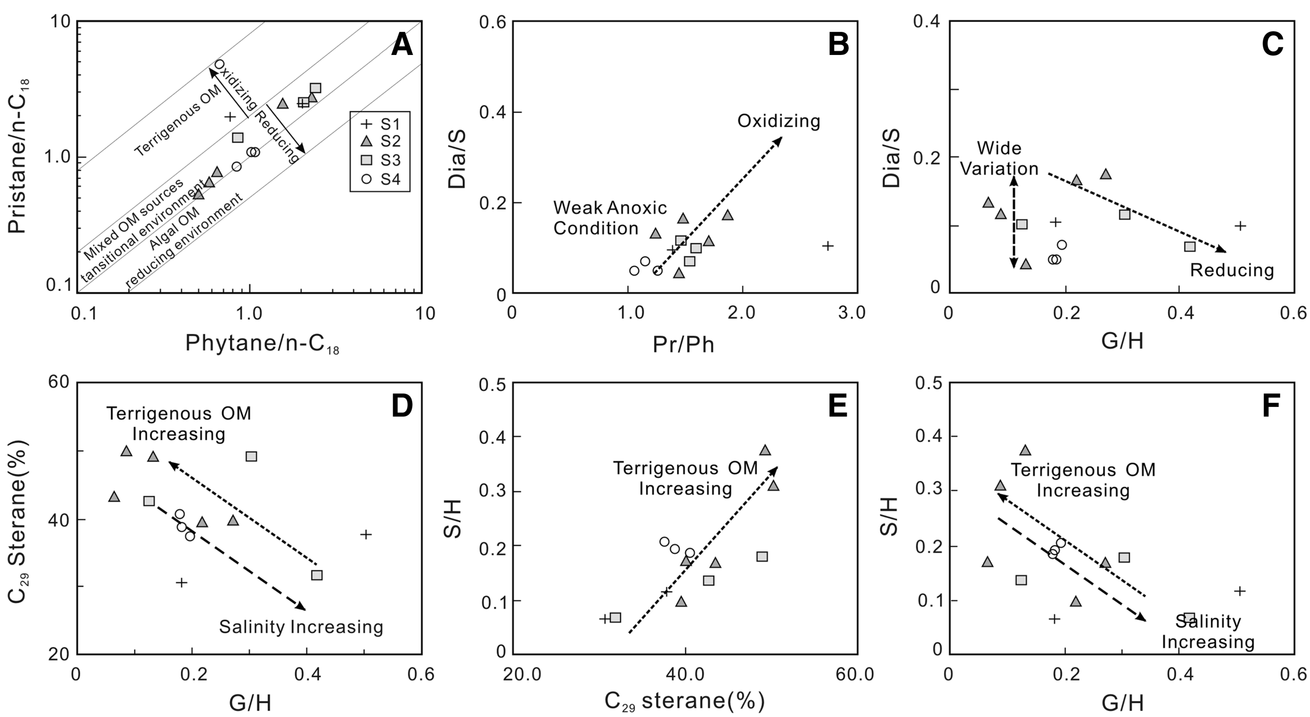

Fig. 4 Correlation between various biomarker parameters reflecting organic matter input and/or depositional environments in the K_1g^1 member. *Abbreviations* for biomarker parameters are explained in Fig. 2

organic matter in Sects. 1 and 3 are dominantly type II and mainly type III with some type II for Sects. 2 and 4.

A clear positive linear correlation between HI and TOC was observed in Fig. 3b, suggesting that the abundance and type of organic matter seem to be controlled by the same factors.

3.2 Depositional environments

Pristane and phytane were identified in all samples from the K_1g^1 member. The Pr/Ph ratio is often used as a redox indicator of depositional environment (Didyk et al. 1978) but with limitations (Peters et al. 2005). The Pr/Ph ratio for the K_1g^1 member ranges from 1.06 to 1.88 with an average of 1.43, except for one sample (2.73) in Sect. 1 (Fig. 4b), which falls in the range considered to be poorly indicative of redox conditions (Peters et al. 2005).

The ratio of diasteranes to steranes is one of the indicators for redox conditions of depositional conditions, as oxic conditions are favorable for diasterane formation. A high diasterane/sterane (Dia/S) ratio (calculated as [total $C_{27}-C_{29}$ diasteranes]/[total $C_{27}-C_{29}$ steranes]) often suggests an oxic depositional condition when samples are at a comparable level of thermal maturity (Peters et al.

2005). Because samples from the K_1g^1 member have similar Tmax (maturity) and lithology, the diasterane/sterane ratio can be interpreted as an indication of redox conditions. As shown in Fig. 4b, the diasterane/sterane ratio is low and varies from 0.04 to 0.18 (average 0.10), suggesting reducing conditions.

Pr/n-C_{17} and Ph/n-C_{18} ratios have been widely used as indicators of depositional environment and organic matter input (Peters et al. 1999; Hanson et al. 2000; Duan et al. 2008). On the Pr/n-C_{17} vs. Ph/n-C_{18} plot, samples from different source rock intervals all fall in the mixed organic matter sources transitional environment fields (Fig. 4a).

Gammacerane was detected in all samples in the K_1g^1 member and shows similar variation as that of the TOC contents, with its gammacerane index (gammacerane/C_{30} 17α(H),21β(H) hopane) ranging between 0.07 and 0.50 with an average of 0.22 (Figs. 2, 5). Gammacerane is widely considered to form by reduction of tetrahymanol in bacterivorous ciliates, which occurs at the interface between oxic and anoxic zones in stratified water columns (Sinninghe Damsté et al. 1995). An abundance of gammacerane is usually considered to indicate the presence of stratified water columns (Sinninghe Damsté et al. 1995; Peters et al. 2005). Although hypersalinity and temperature can both result in a stratified water column, a high abundance of gammacerane is mostly found in evaporitic or high salinity environments (Fu et al. 1990; Ritts et al. 1999; Hanson et al. 2000; Summons et al. 2008). As shown in Fig. 4c, a high abundance of gammacerane combined with low Dia/S suggests stable salinity-stratified water column with anoxic bottom water conditions, while a low abundance of gammacerane with variable Dia/S suggests that unstable stratified water column occurred due to a temperature gradient (e.g. Bohacs et al. 2000), which resulted in variable redox conditions in low salinity water. This is evidenced by the fact the alternating beds mudstones and silty mudstones occurred in Sect. 2, revealing alternating deposition environments. Lamellar mudstones and mudstones occurred in Sect. 3, suggesting relatively stable deposition environments.

3.3 Organic matter sources

The compositions of organic matter in the K_1g^1 member are mainly algae-bacterial organic matter (AB, including alginite and mineral bituminite), varying from 30.7 % to 90.7 % (average 62.9 %). The relative content of terrigenous high plant organic matter (TPH, including vitrinite, inertinite, sporinite, cutinite, and exinite debris) is variable but usually low (Fig. 2). This suggests important contributions from algae, planktons and bacteria, and variable terrigenous high plant input (Powell et al. 1990; Xiao and Jin 1990; Ercegovac and Kostić 2006).

Sterane distribution has the potential to reflect the primary producer in marine and lacustrine systems (e.g. Volkman 1986; Knoll et al. 2007; Sepúlveda et al. 2009; Hao et al. 2011). While C_{28} steranes are associated with specific phytoplankton types (e.g. Volkman et al. 1998), C_{27} steranes mainly originate from phytoplankton and metazoa, and C_{29} steranes mainly derive from terrigenous higher plants (e.g. Volkman 1986; Huang and Meinschein 1979). $C_{27–29}$ steranes/$C_{27–35}$ hopanes (S/H) are also used to indicate input of eukaryotic (mainly algae and high plants) versus prokaryotic (bacteria) organisms (e.g. Peters and Moldowan 1993; Hanson et al. 2000). As shown in Fig. 4d–f, S/H increases with the rise of C_{29} sterane abundance, and C_{29} sterane content and S/H increase with the decrease of G/H, suggesting high S/H was caused by terrigenous higher plant contribution and that high salinity was favorable for bacterial mats, resulting in low S/H.

The $\delta^{13}C$ of total organic matter ranges from −33.0 ‰ to −24.6 ‰ (Fig. 2), but it shows significantly higher values in Sect. 2 compared to the other sections. As shown in the Fig. 6, the $\delta^{13}C$ value increases with the increasing C_{29} sterane abundance, suggesting terrigenous organic matters have high $\delta^{13}C$ values than algal-sourced organic matter in Cretaceous, which is consistent with the previous studies (Dumitrescu and Brassell 2006; Gröcke et al. 2003; Yans et al. 2010).

Samples from Sect. 3 have high AB content and low THP content, and combined with low abundance of C_{29} sterane, low S/H, and light $\delta^{13}C$, they suggest important contributions from algae, phytoplankton and bacteria (Fig. 2). In contrast, samples from Sects. 2 and 4 have relatively high THP content and low AB content, and combined with high abundance of C_{29} sterane, high S/H and heavy $\delta^{13}C$, this indicates an important contribution from terrigenous high plants (Fig. 2).

3.4 Mechanism of organic matter accumulation in response to the climate

Studies of the fossil plants and sediments show that the lacustrine source rocks associated with the Xiagou Formation were deposited in the arid and semi-arid climate (Pan et al. 2006; Deng and Lu 2008). Both the accumulation and type of organic matter are controlled by the same factors (Fig. 3a). Although the number of samples subjected to the maceral, isotopic, and molecular analysis is limited, TOC and HI have obvious correlations with organic matter biomarker parameters, depositional environment biomarker parameters, maceral and carbon isotopic composition (Fig. 6). Therefore, TOC and HI have the potential to reflect the enrichment and source input of organic matter, and the depositional environments that

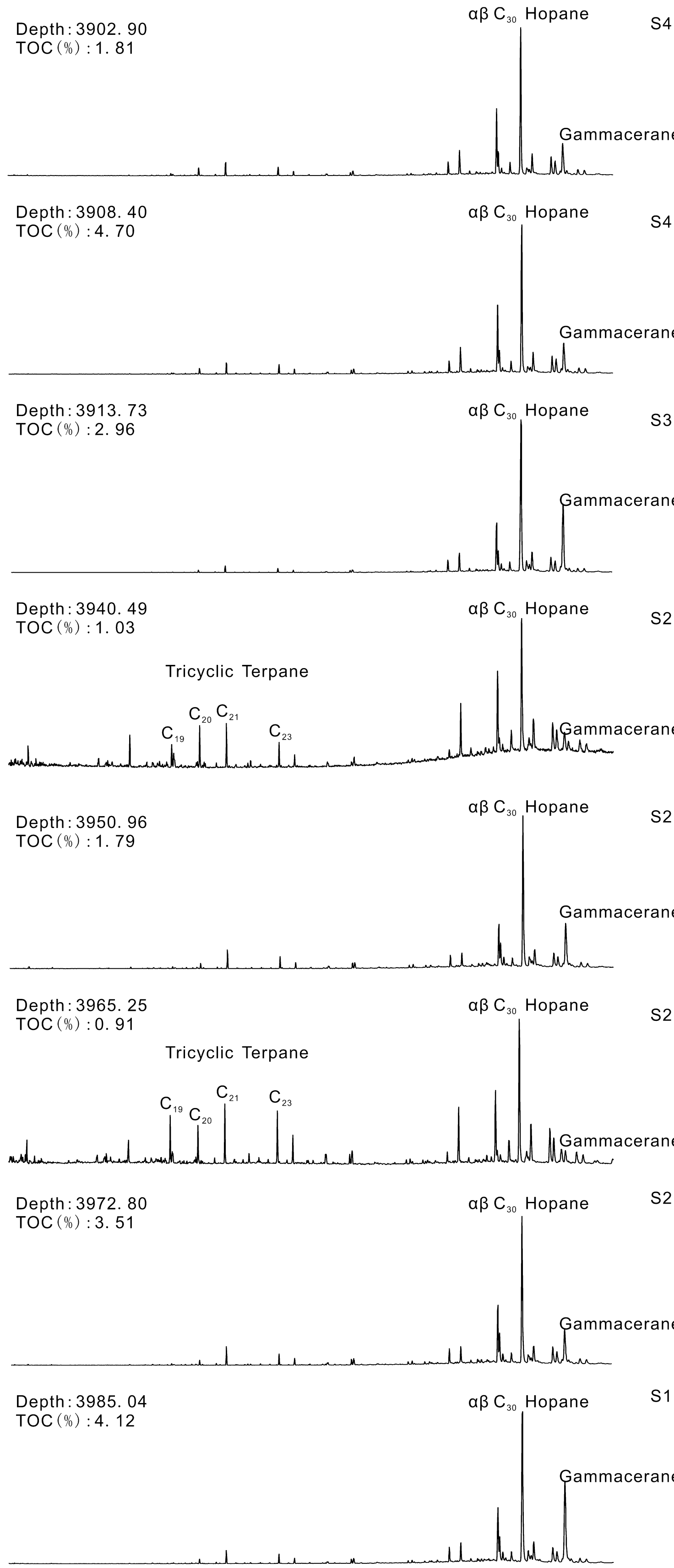

Fig. 5 Representative m/z 191 mass chromatograms showing variation in the composition of terpanes through the Chang 2-2 borehole profile

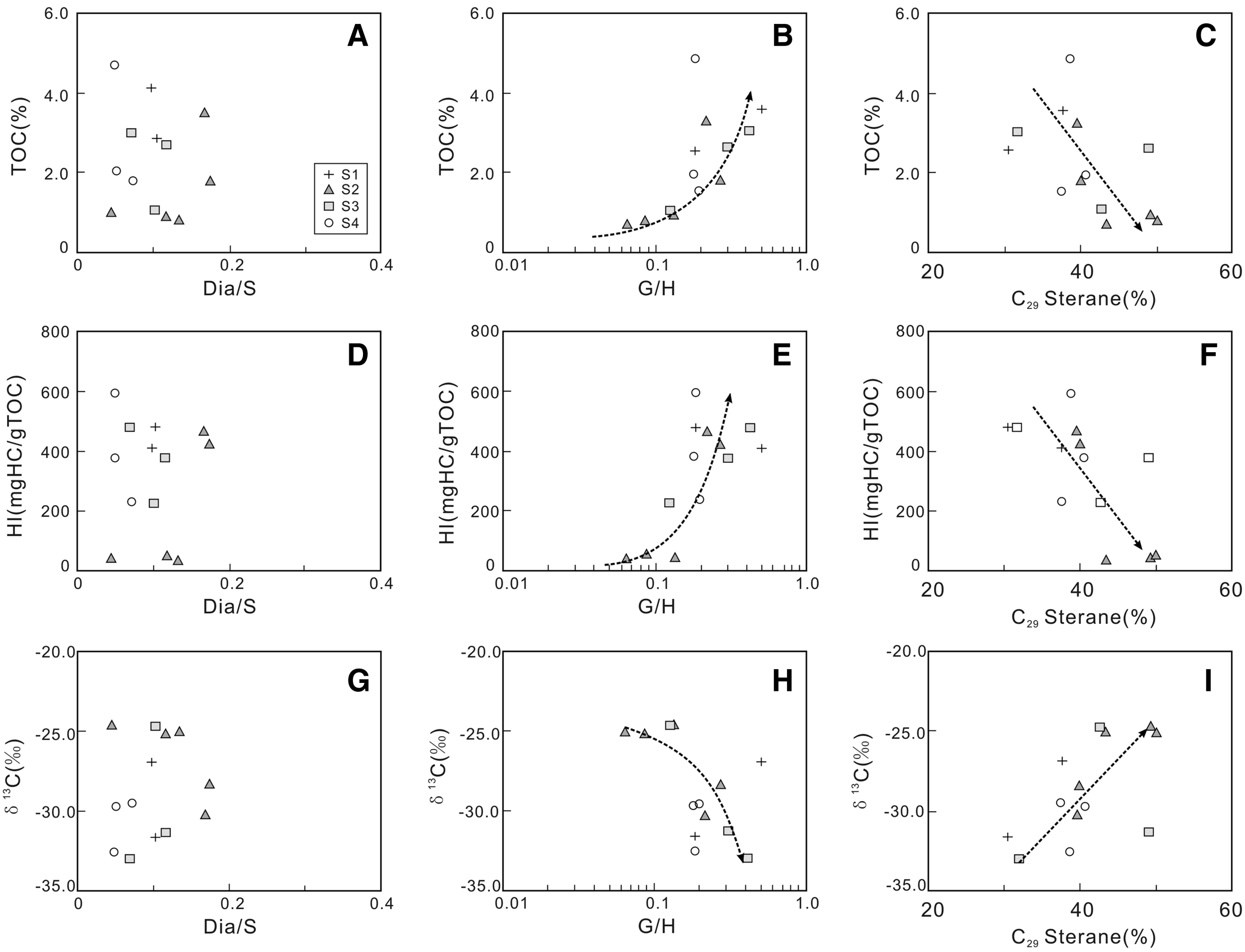

Fig. 6 Correlation between TOC, HI, kerogen $\delta^{13}C$ and maceral compositions for the K_1g^1 member samples, showing the controlling factors of accumulation and type of organic matter. *Abbreviations* for biomarker parameters are explained in Fig. 2

form in response to the evolution of the paleoclimate and abundant samples subjected to Rock–Eval analysis were effectively used to analyze the mechanism of lacustrine organic matter combined with limited maceral, molecular, and isotopic analysis.

TOC, HI and $\delta^{13}C$ are poorly correlated with Dia/S (Fig. 6a, d. g) but well correlated with G/H (Fig. 6b, e, h) and C_{29} sterane abundance (Fig. 6c, f, i), which seems to suggest that the enrichment of organic matter was more controlled by the salinity and organic matter source than by redox conditions in the K_1g^1 member. The salinity of lake has an effect on the growth of bacteria, algae, planktons, and high plants, and high salinity seems to be favorable for high primary productivity of algae, planktons, and bacteria (Fig. 4d, f). Some samples in Sects. 2 and 4 with low G/H and low Dia/S have low TOC, suggesting that the fresh lakes with stratified water (low G/H and low Dia/S) resulting from temperature gradient (e.g. Bohacs et al. 2000) had good organic matter preservation conditions. They also had low primary productivity of algae, planktons, and bacteria in the fresh waters and received significant terrigenous high plants input, resulting in low TOC, heavy $\delta^{13}C$, and low content of ABOM in Sects. 2 and 4 (Fig. 2). Comparatively, the relatively high salinity water had both high primary productivity of algae, planktons, and bacteria and good organic matter preservation conditions (low Dia/S) in Sects. 1 and 3, resulting in high TOC, light $\delta^{13}C$, and high ABOM content in Sects. 1 and 3 (Fig. 2).

Both the primary productivity and salinity of lakes can be well interpreted on the paleoclimate evolution. In the relatively high temperature and arid climate, lakes had high salinity, high primary productivity of algae, planktons, and bacteria, and good organic matter preservation conditions, resulting in high TOC, light $\delta^{13}C$ and high ABOM content (Sects. 1 and 3 in Fig. 2). Comparatively, in the relatively low temperature and wet climate, lakes had low salinity, low primary productivity of algae, plankton, and bacteria, and they received significant terrigenous high plants input, thus resulting in low TOC, heavy $\delta^{13}C$, and low content of ABOM (Sects. 2 and 4 in Fig. 2).

4 Conclusions

This analysis demonstrates that variations of TOC, HI, maceral composition, and carbon isotopic composition of total organic matter and biomarker parameters for samples from the K_1g^1 member in the Chang 2-2 borehole mainly reflect the enrichment and source inputs of organic matter and depositional conditions in response to the climate evolution. Sections 1 and 3 with high TOC, HI, algae-bacterial organic matter content and light $\delta^{13}C$ deposited in high salinity lakes with high primary productivity of algae, planktons and bacteria and good organic matter preservation conditions in the relatively high temperature and arid climate. Sections 2 and 4 with low TOC, HI, and algae-bacterial organic matter content, and heavy $\delta^{13}C$ deposited in less saline lakes with low primary productivity of algae, planktons and bacteria in relatively wet climate. The enrichment and type of organic matter was controlled by the primary productivity and preservation conditions in response to the climate evolution during the period of the K_1g^1 member deposition in the Jiuquan Basin.

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